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Reply to Office Action of October 24, 2003

REMARKS

Claims 3, 7-9 and 10-18 and new Claims 19 and 20 are active in the case.

Reconsideration is respectfully requested.

Applicants' representative wishes to thank Examiners Ridley and Johnson for the helpful and courteous interview of December 3, 2003. As a result of the discussion it is believed that the issues in the case have been clarified and that the prosecution of the application has been materially advanced.

The present invention relates to a method for the production of synthesis gas.

Claim Objection

The objection to Claims 3 and 17 is believed obviated by the amendment to Claim 3 by which the dependency of the claim is now clearly indicated as Claim 10. Accordingly, withdrawal of the objection to Claims 3 and 17 is respectfully requested.

Claim Amendments

Minor amendments have been made to each of Claims 3 (as indicated and discussed above) and 17, as well as other claims. Claim 10, in particular, has been amended to eliminate the term "catalyst" therefrom because the hexavalent chromium oxide functions as an oxidant, and has been further amended to correct an error in the deletion of another metal oxide from original Claim 1 that functions by exothermically promoting the oxidation of the light hydrocarbon to synthesis gas. Still further, original Claim 2 has been reinstated in the form of new Claim 19. Finally, new Claim 20 has been presented which is similar to Claim 10 except

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that it recites that the hydrocarbon stream is solely a light hydrocarbon gas that is converted to synthesis gas as is indeed supported, for example, by page 16, last full paragraph and the paragraph bridging pages 16 and 17, which shows the use of CrO_3 in reaction with methane alone to form synthesis gas product. The amendments do not introduce new matter into the case and entry of the amendments is respectfully requested.

Claim Rejection, 35 USC 112

The issue raised with respect to Claim 18 is not, in fact, believed to be an issue. It is pointed out that while Claim 18 depends on Claim 15 and Claim 15 depends on Claim 14, Claim 14 depends on Claim 10 which is the claim that mentions a catalyst containing chromium trioxide and an alkali or alkaline earth metal. Accordingly, ultimately, it is Claim 10 that provides antecedent basis for the catalyst of Claim 18. Thus, the indefiniteness issue that has been raised is believed overcome and withdrawal of the same is respectfully requested.

Invention

As stated previously in the record, the present invention is a process of producing synthesis gas by partially oxidizing or autothermally reforming a light hydrocarbon gas with a hexavalent chromium oxide, supported on an inert carrier and modified with an alkali or alkaline earth metal, as the light hydrocarbon contacts the hexavalent chromium oxide and extracts oxygen therefrom thereby being partially reduced, and metal oxides that are capable of autonomously sustaining the catalytic partial oxidation reaction by means of redox cycles.

Using reactions as a description of the present invention, the objective reaction of the

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present process is the partial oxidation of a light hydrocarbon to synthesis gas as follows:



(methane is an example of a light hydrocarbon)

The oxygen for the partial oxidation reaction above is supplied by the reduction of hexavalent chromium oxide to trivalent chromium oxide as follows:



Both reactions occur as a light hydrocarbon gas, such as methane, is passed into a fluidized bed within an oxidation reactor containing a metal oxide material, a primary component of which is CrO_3 .

Claim Rejection over Prior Art

Claims 7-13 stand rejected based on 35 USC 102(b) or 35 USC 103(a) as anticipated by or as obvious over Dickinson, U.S. Patent 2,602,809. This ground of rejection is respectfully traversed.

The Examiner comments in the Official Action that the Dickinson patent teaches the partial oxidation or autothermal reforming of a light hydrocarbon to synthesis gas. To the contrary, the patent teaches **no such method**. As is clear from the disclosure of the paragraph bridging columns 2 and 3 of the patent, a solid-carbon containing material such as coal or coke is heated in a reactor with a fluidized metal oxide such as magnetite (Fe_3O_4) or hematite (Fe_2O_3). Gasification occurs as the carbon material is oxidized and the preferred gaseous material is CO

(In Fig. 1 of the patent, the oxidation of a solid carbonaceous material to gaseous product occurs in reactor 13.) As described at the top of column 3, the reduced metal oxide obtained from the reactor is regenerated (oxidized) to its original oxidation state by treatment with steam or air. The regenerated metal oxide is then reused to form more gaseous carbon oxide product. Clearly, this reaction is **not** the partial oxidation or autothermal reforming of a light hydrocarbon to synthesis gas.

Another aspect of the process described in the patent is that, **if desired**, hydrogen can be produced to blend with the carbon oxide product produced in reactor 13 as described at column 3, lines 11-41. In this aspect of the disclosed process, a portion of the spent, i.e., reduced, catalyst obtained from the oxidation reactor 13 is passed to oxidation reactor 24 (In Fig. 1) where the reduced metal oxide is contacted with steam. The products of this reaction are hydrogen and reoxidized metal oxide. The reoxidized metal oxide is then passed back to the coal or coke gasification unit 13 and the hydrogen product is combined with the carbon monoxide passed from the reactor 13, if desired to form synthesis gas. Clearly, the oxidation reaction which produces hydrogen as the sole gaseous product is also **not** the partial oxidation or autothermal reforming of a light hydrocarbon to synthesis gas. In fact, there is no disclosure anywhere in the patent of passing a light hydrocarbon gaseous material into a reactor where a fully oxidized metal oxide reactant is present to produce a mixture of CO and H₂ (synthesis gas)! If a synthesis gas product is to be produced by the process disclosed in the patent, it is accomplished only by combining H₂ and CO obtained from separate sources or separate reactions and combined in the relative amounts desired.

As to the matter of the disclosure of CrO₃ as a metal oxide as a reactant with the solid

carbonaceous starting material for the oxidation reaction within reactor 13, no such disclosure is found in the patent. The preferred metal oxide reactants for use in the oxidation reactor are hematite and magnetite. Other possible metal oxides include any one of the several metal oxides mentioned at column 9, lines 39-49 of the patent. None of the several metal oxides mentioned is specifically CrO_3 . Chromium oxides (specifically unidentified) are mentioned, but here there is not the slightest hint of employing CrO_3 as a source of oxygen for the oxidation of a light hydrocarbon. In this regard the Examiner and her supervisor took the position during the interview that a chromium oxide for the process of the reference could be CrO_3 and that the oxide would interact with methane gas, if such were to be the gas selected amongst those mentioned at lines 29-32 of column 4 of the patent, for passage into gasification reactor 13 to produce synthesis gas. However, applicants submit that this is an un-warranted stretch of the teachings of Dickinson. Dickinson clearly states at column 4, lines 29 *et seq* that the gas selected, whether steam, CO_2 , methane or recycle gas, is done so merely as a gas that fluidizes or carries particulate matter, particularly in the reaction zone. Accordingly, methane gas is not taught by the patent in any way as participating in the production of synthesis gas in reactor 13. In fact, it is clear that such could not be the case, because the several gases mentioned are equivalent in their intended roles as fluidizing gases, **not reactants**. (New Claim 20 is clearly distinct from Dickinson because it specifies that a light hydrocarbon gas is the only hydrocarbon that is partially oxidized to synthesis gas.) Moreover, there is absolutely nothing in the reference which would lead one of skill in the art to specifically use CrO_3 in the reaction zone to interact with methane, disclosed only as useful as a fluidizing gas, for the production of synthesis gas, which is not taught as a reaction product of reactor 13.

Applicants also refer to the following experiment as described which employs an unsupported chromium oxide catalyst as disclosed in Dickinson in the oxidation reaction of the invention. The experiment is as follows:

In the apparatus described and shown in the present application, wherein the oxidation reactor has an internal diameter of 3.5 cm and the regenerator has an internal diameter of 5 cm, a 4900 gram quantity of solid particulate chromium oxide catalyst material having an average particle diameter equal to 70 μm as placed. The catalyst was pure, unsupported Cr_2O_3 . Once the catalyst was fluidized in the two containers, the following distribution of solid material was obtained:

- a) 2700 gr in the regenerator;
- b) 1900 gr in the reactor;
- c) the remaining 300 gr equally distributed in the conveying lines.

The conveyance of the solid catalyst material between the reactor and the generator was regulated, so as to have in both directions, a flow rate equal to 5 kg/h of solid material. The regenerator was kept at a temperature of 700° C and an internal pressure of 2 Mpa. Preheated air at a flow rate of 650 Nl/h was fed to the regenerator. The effluent gas exiting the regenerator had a flow rate of 649.7 Nl/h which indicates that only a small amount of oxygen had been absorbed and consumed on the solid surface of the catalyst. Chemical analysis confirmed that only about 0.1 % of the Cr_2O_3 had been transformed to CrO_3 .

The catalyst material oxidized in the regenerator was pneumatically conveyed to the top of the reactor at a flow rate of 5 kg/h of solid material. Methane gas was also passed into the reactor at a flow rate of 172 Nl/h under a pressure of 20 atm and a temperature of 900° C. An

effluent gas stream at a flow rate of 173.2 Nl/h was taken from the reactor. The gas was composed mainly of methane with had minor amounts of H₂, CO, CO₂ and H₂O vapor. Chemical analysis confirmed that the small amount of CrO₃ originally present in the catalyst was re-transformed into Cr₂O₃.

Clearly, Dickinson fails to either teach or suggest the present invention, and withdrawal of the rejection is respectfully requested.

Claims 14 and 15 stand rejected based on 35 USC 103(a) as obvious over Dickinson, U.S. Patent 2,602,809. This ground of rejection is respectfully traversed.

Beyond the fact that Claims 14 and 15 ultimately depend on Claim 10 which is clearly patentably distinguished over the Dickinson patent, the specific subject matter of Claims 14 and 15 is not taught anywhere in the reference. Moreover, the simple disclosure of alumina as a support at column 10, line 44 is hardly a disclosure of the specific microspheroidal alumina of Claims 14 and 15. Many different types of alumina can be produced depending upon the methods used for synthesis. There is absolutely no teaching or suggestion of a method of syntheses in the reference by which microspheroidal alumina can be produced. Clearly, the subject matter of Claims 14 and 15 is not taught anywhere in the reference and therefore withdrawal of the rejection of the claims is respectfully requested.

Claims 16 and 18 stand rejected based on 35 USC 103(a) as obvious over Dickinson, U.S. Patent 2,602,809 in view of Kmecak et al, U. S. Patent 3,965,252. This ground of rejection is respectfully traversed.

As to the Kmecak et al patent, the same is irrelevant to the present invention because it discloses an entirely different type of reaction from, not only the present invention, but also that

in Dickinson. Kmecak et al discloses a reaction in which a light hydrocarbon such as ethane, propane or the like, is reacted in the presence of a Group VIII noble metal or a Group VIB metal and a promoter of the likes of a Group I metal is degraded, thereby producing hydrogen gas. Synthesis gas is not produced by the process of the reference. Further, there is no teaching or suggestion of a process by which a light hydrocarbon gas is subjected to a metal oxide catalyst which promotes the partial oxidation of the light hydrocarbon, thereby degrading the same to produce a carbon monoxide/hydrogen gas mixture. Still further, the specific CrO_3 reactant of the present process is nowhere shown or suggested. Accordingly, when this reference is combined with Dickinson, there is clearly no teaching or suggestion of a process of producing synthesis gas by the partial oxidation or autothermal reforming of a light hydrocarbon to synthesis gas in the presence of at least CrO_3 mixed with another metal oxide and an alkali metal promoter. Accordingly, the cited ground of rejection is believed overcome and withdrawal of the rejection is respectfully requested.

Claims 7-13 stand rejected based on 35 USC 103(a) as obvious over Dickinson, U.S. Patent 2,602,809 in view of Rooks et al, U. S. Patent 4,367,160 or Cole, U. S. Patent 6,007,699. This ground of rejection is respectfully traversed.

The relevancy of the cited Rooks et al patent to either the Dickinson patent or the present invention is not seen. Contrary to the Examiner's statement at page 7, lines 5-8 of the last paragraph of the Office Action, Rooks et al nowhere shows or suggests that hexavalent chromium oxide can be used for the production of synthesis gas. (Reference is apparently made to the disclosure of iron chromite as an oxidant. However, such is not CrO_3 , but is a combination of iron oxide and trivalent chromium oxide.) While Rooks et al mentions in the abstract that

synthesis gas production is an aspect of the disclosure, there is no disclosure of a light hydrocarbon as a source material. Carbon-containing materials mentioned in the patent are those in the last full paragraph of column 3. No light hydrocarbon gas is mentioned here. Rather, Rooks et al teaches in one of the several aspects of the invention as described in the paragraph of columns 1 and 2, that a carbon-containing material, such as one of those taught at the bottom of column 3, none of which is a light hydrocarbon gas, is heated in the presence of a metal oxide (oxidant) which releases oxygen and functions as a heat transfer agent. The heated oxidant is at least partially reduced with synthesis gas to form steam and carbon dioxide. In this process the objective is to convert the solid or heavy liquid starting material to carbon monoxide (see column 3, lines 4-9). Clearly, the reference does not teach the preparation of synthesis gas as a product, and certainly does not teach or suggest the select use of CrO₃ as a source of oxygen for the conversion of a light hydrocarbon to a synthesis gas product. Accordingly, the Rooks et al disclosure does not bring the Dickinson patent closer to the present invention. Indeed, the combination of the two patents is difficult because it is not at all clear how a process in which a metal oxide oxidant that is partially reduced by synthesis gas, when used to gasify carbon-containing material to carbon monoxide, can be adapted for use in a process where a fully oxidized metal oxide is used to provide oxygen for the partial oxidation of solid carbonaceous material to CO.

As to the Cole patent, the same is irrelevant to the present invention, because it nowhere teaches or suggests a method of converting a light hydrocarbon to synthesis gas. Rather, Cole is concerned with a process of oxidizing fuel and transferring the heat produced to a particular use in a combustion system. Such has nothing to do with synthesis gas production. The

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Examiner refers to the patent as teaching Cr (VI) as a catalyst. However, the metal ions mentioned in the paragraph of columns 8 and 9 only mentions Cr(III)/Cr(IV) oxide as a combustion catalyst for the oxidation of a fuel. Such is not germane to synthesis gas production.

Claims 14 and 15 stand rejected based on 35 USC 103(a) as obvious over Dickinson, U.S. Patent 2,602,809 in view of Rooks et al, U. S. Patent 4,367,160 or Cole, U. S. Patent 6,007,699. This ground of rejection is respectfully traversed.

Applicants retain their statements as set forth above concerning the distinction of the subject matter of Claims 14 and 15, as they ultimately depend on Claim 10, which, in turn, is clearly patentably distinguished over the Dickinson patent. The specific subject matter of Claims 14 and 15 is not taught anywhere in the reference. Moreover, the simple disclosure of alumina as a support at column 10, line 44 of Dickinson is hardly a disclosure of the specific microspheroidal alumina of Claims 14 and 15. Moreover, neither to Rooks et al patent nor the Cole patent teaches anything other than that alumina (generally) is a common catalyst support. Such disclosures does not lead one of skill in the art to the specific use of a microspheroidal alumina as a catalyst support, not only generally, but also for a metal oxide that functions as an oxidizing agent for a light hydrocarbon. Accordingly, withdrawal of the rejection of the claims is respectfully requested.

Claims 16 and 18 stand rejected based on 35 USC 103(a) as obvious over Dickinson, U.S. Patent 2,602,809 in view of Rooks et al, U. S. Patent 4,367,160 or Cole, U. S. Patent 6,007,699. This ground of rejection is respectfully traversed.

There is absolutely no teaching in either of the Rooks et al patent or the Cole patent of the fabrication of a metal oxide material based on CrO₃ that is particularly useful as a source of

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oxygen for reaction with a light hydrocarbon gas, particularly where the metal oxide in addition is mixed with potassium and a metal oxide system where CrO₃ and potassium oxide are mixed in particular relative amounts. Accordingly, the two references do not improve upon the deficiencies of Dickinson and withdrawal of the rejection is respectfully requested.

It is now believed that the application is in proper condition for allowance. Early notice to this effect is earnestly solicited.

Respectfully submitted,

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